Clemson PI expands knowledge of high temperature sorption behaviors for used nuclear fuel storage

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With continued uncertainty about used nuclear fuel storage, more research is needed to make informed decisions. Brian Powell, a Clemson University PI, and his research team hope to add real scientific data to the discussion.

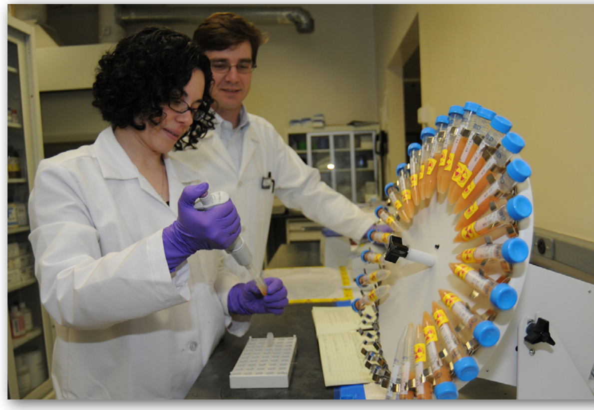
One of the major used fuel storage challenges is understanding how radioactive contaminants move and change over time. Some minerals and materials absorb radioactive materials, preventing contaminants from leaching into the environment outside of the storage facility.

Chemical reactions caused by different environmental factors, such as exposure to oxygen or carbon dioxide, can change the actinide or radioisotopes' state, or species, making it behave differently than it originally would. Different species will exhibit different sorption behavior and thus different mobility in the environment. Traditionally, empirical constants have been used to predict movement of radionuclides in the environment. This technique only provides results for the conditions under which the empirical constants were measured meaning it cannot account for the changing conditions inside of a changing geologic environment.

That's where Powell and his team take over.

"We are trying to gain a better understanding of transuranic elements and how they are going to behave," said Powell. "We want to get away from the empirical models and incorporate specific chemical reactions into fate and transport models."

The team's novel approach focuses on creating an accurate dataset for use in predicting the specific chemical species present in aqueous and solid phases within the environment. A unique collaboration combines Clemson's expertise in actinide envrionmental chemistry with Lawrence Berkeley National Laboratory's expertise in isothermal titration calorimetry and chemical thremo-dynamics and University of California, Berkeley's expertise in x-ray absorption spectroscopy to identify the different chemical species involved.



Powell supervises PhD student, Amy Hixon, as she withdraws a mineral suspension containing trace level plutonium. The sample will be analyzed to determine the amount of plutonium sorbing to the mineral.

"This was a natural marriage between calorimetry measurements, x-ray absorption spectroscopy and sorption experiments," said Powell. "Coupling the three technical approaches creates accurate data to prove the principle."

These techniques, when combined, create a fundamental understanding of the reactions occurring during sorption of an actinide to a mineral surface which are quantified with thermochemical constants rather than empirical constants. By using these thermo-chemical constants, models can consider all possible chemical species, resulting in a more robust, versatile, and technically consistent model.

The idea for the research was hatched seven years ago, while Powell was working for now collaborator, Dr. Linfeng Rao at Lawrence Berkeley National Laboratory, who is an expert in actinide thermodynamic studies and calorimetry measurements.

"These early discussions really laid the foundation for the theoretical background for these sorption entropy and enthalpy measurements, which is the key component of the research," said Powell.

The compilation of project data will provide a vast dataset to understanding thermochemical reactions which are able to predict actinide speciation under different conditions.

Databases listing energies involved in chemical reactions are not a new concept. The International Nuclear Energy Agency has been compiling data for aqueous actinides species for several years. Powell and his team hope to create a similar database for speciation on mineral surfaces, improving modeling and simulation accuracy in the area.

Powell emphasizes that the database can be useful for any disposal site, not just a repository. He hopes that his research will inform decision makers and open up candid discussion about viable fuel cycle options.

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